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Margaritis, N.; Blum, Ludger; Batfalsky, Peter; Ceschini, Sergio; Fang, Q.; Federmann, Dirk; Kroemer, Joachim; Menzler, Norbert H.; Peters, Roland; Steinberger-Wilckens, Robert

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N. Margaritis[a], L. Blum[b], P. Batfalsky[a], D. Bohmann[d], S. Cechini[c], Q. Fang[b], D. Federmann[a], J. Kroemer[d], Ro. Peters[b], R. Steinberger-Wilckens[e]

[a] Forschungszentrum Jülich GmbH, 52425 Jülich, Germany, Central Institute for Engineering, Electronics and Analytics (ZEA)
[b] Forschungszentrum Jülich GmbH, 52425 Jülich, Germany, Institute of Energy and Climate Research (IEK)
[c] SOLIDpower S.p.a., 38017 Mezzolombardo (TN), Italy
[d] Borit NV, 2440 Geel, Belgium
[e] University of Birmingham, School of Chemical Engineering, College of Engineering and Physical Sciences, UK

Lightweight SOFC stacks are currently being developed especially for automotive applications such as APU and for portable devices. Within the EU funded project MMLCR=SOFC the Jülich lightweight so-called CS-design was improved concerning better suitability for glass sealing, reduced manufacturing effort and increased power. Based on modelling in combination with manufacturing experience, test results, and post-test analysis substantial changes of the previous CS-design were made. The manufacturing of single parts, particularly due to the improved design of sheet metal interconnects, as well as the assembling processes are suitable for low-cost mass manufacturing. The novel decal concept of glass-ceramic sealant screen printed on foil in order to produce green tapes is used for joining the stack layers offering an enormous potential for cost savings in industrial assembly process. First stack tests with the new CS^{V}–design showed a comparable electrochemical performance to the previous CS^{IV} design having at the same time a better thermo-mechanical behavior.

Introduction

Lightweight SOFC stacks are currently being developed for stationary applications such as residential CHP units, for automotive applications such as APU and for portable devices. They allow electrical efficiencies of up to 60%, high fuel flexibility, being able to operate on syngas from Diesel reforming as well as on LPG, methane or hydrogen, and they are promising low costs due to greatly reduced amounts of steel interconnect material and cheap manufacturing processes.

The requirements for thermal cycling in lightweight stacks are high, since start-up times for vehicles of most kinds (road and rail vehicles, even aircraft and ships) are short compared to those for stationary power generation. The same applies to portable devices which are expected to be operational within very short time intervals. But also for stationary applications rapid start-up will be an advantage since the SOFC system can be operated more closely in accordance with the electrical and thermal loads of the customer. It is therefore essential that lightweight stacks have excellent thermo-mechanical properties. This requires a specific stack design that compensates mechanical stresses from temperature gradients in space and time. Simultaneously the cost pressure on mobile and portable applications is high. It is therefore important to manufacture stacks to high quality standards and implement as many automated precision assembly processes as possible in an approach of parallel improvement of quality and lowering of manufacturing cost.

Development of light weight design SOFC

First Designs

The light weight design from JUELICH, called CS design, had to be improved to become more stable under thermomechanical stress, taking into account results from the former tests [1]. Tests with the first stacks of the next generation CS^{II} design revealed that the contact area on the cathode side is quite small because of the sinoidal shaped channel structure, which is the optimum for the forming of plates. Tests using hydroforming instead of classical stamping showed that it is possible to manufacture channel structures with a more flat surface. This was implemented in the CS^{IV} design (see Fig. 1).

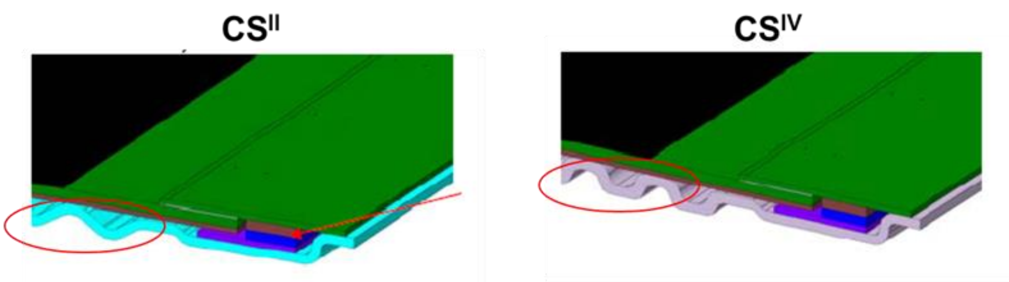

Figure 1. Improvements from Design CS^{II} to CS^{IV}: Improved contact with flat channel structure in CS^{IV} design.

To improve the mechanical robustness several swages (L and U-seam) were implemented in CS^{III} design. This should make the cassette stiffer and reduce the stress in the glass sealing. Assembly trials and first stack tests revealed that this design was among others much too sensitive to manufacturing tolerances. Therefore only the U-seam was implemented into the CS^{IV} design (see Fig. 2), which promised higher robustness without showing all the problems of CS^{III}.

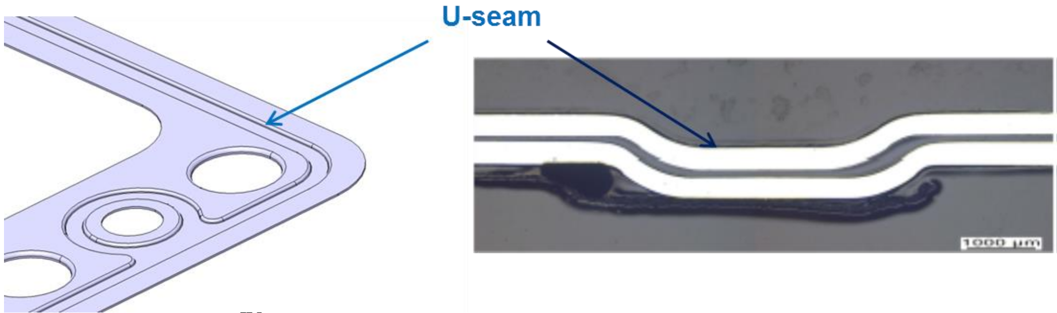

Figure 2. Design CS^{IV} with swage (or U-seam), to improve the mechanical robustness during thermal cycling.

This CS^{IV} design was used for first stacks to be manufactured within the current EC-project and Borit was manufacturing tools and parts based on the design shown in Figures 3 and 4.

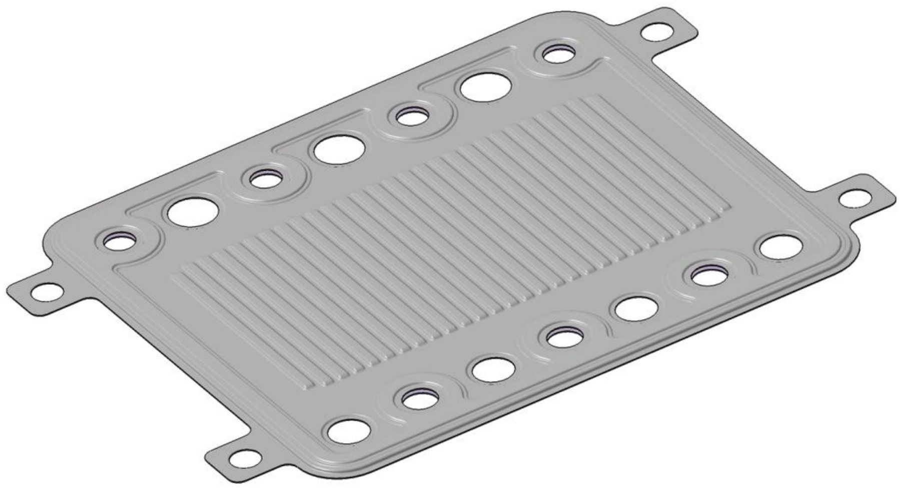
Figure 3. Design CS^{IV}: Cassette after laser welding– tray side.

These stacks showed good electrical performance but the thermo-mechanical behavior was still not good enough. Also during manufacturing several problems occurred because of the small distances between the various manifold openings and between cell sealing area and manifold. A feature implemented in design CSIV which was thought to improve the mechanical robustness – the U-seam as shown in Figure 2 - created a lot of problems during manufacturing and assembly, because the allowed tolerances to avoid short circuit are very small. Therefore FEM calculations were done which revealed that there is no improvement of the mechanical robustness by this U-seam.

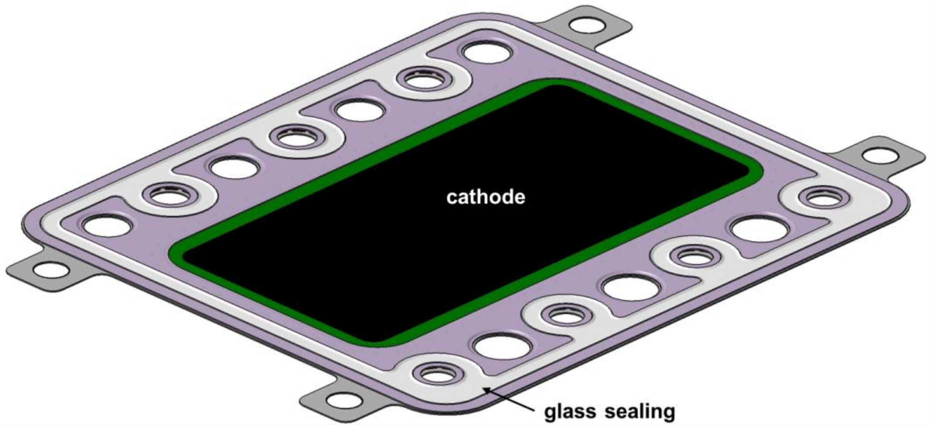

Figure 4. Design CS^{IV}: Cassette – frame side.

Improved Design

Based on these experiences a further improved design was envisaged, mainly addressing the thermomechanical robustness and ease of assembly. Based on testing and modelling results [2, 3, 4, 5] with the JUELICH standard design for stationary applications (so-called F-design) the increase of the sealing area, especially in the manifold array, and the distance between critical locations in the manifold area have been identified as important features. This was applied to the new cassette design, called CS^{V}. While increasing the distance between the feedthroughs automatically the cell area has to be increased too. This results in an increase of the width of the cell from 140 to 180 mm which gives an additional 5 mm between the feedthroughs. An additional issue is the quite small distance from the air feedthroughs to the cell. In this area high temperature gradients create high deformation and so high thermomechanical stress [6]. Therefore the air feedthroughs are shifted towards the outer rim. To keep a reasonable ratio between total area and cell area also the length of the cell is increased from 79 to 100 mm. The drawing of the CS^{V} cassette is shown in Fig. 5.

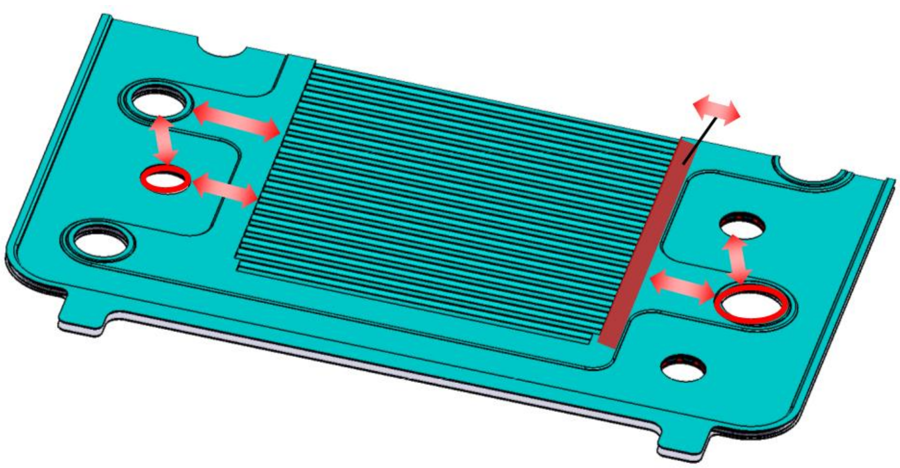

Figure 5. Draft drawing of cassette of design CS^{V} indicating the critical positions which have been enlarged.

Based on this drawing the fluid-dynamic layout concerning in plane flow distribution and from plane to plane flow distribution was performed. To do this first of all the stack parameters had to be determined.

The used operation parameters (for example for a truck APU) are:

- Stack power: 3000 W
- Current density cell: 0.5 A/cm²
- Cell voltage: 0.8 V

Based on a cell area of 137 cm², the number of cells per stack results to 55. The stack will be operated with a mixture of hydrogen and nitrogen of 1:1, which represents the gas coming from a diesel reforming process, and with inlet and outlet temperatures of 600 respectively 800 °C. Based on this stack lay-out data, the gas flow parameters are:

- Air flow per layer: 7.1 g/min (excess air factor = 4.8)
- Fuel flow per layer: 0.92 g/min (fuel utilization u_F = 70%)

Having defined the flow parameters the fluid volume has to be designed. Based on the drawing in Fig. 5 first the cathode side was realized as shown in Fig. 6. The calculation of the pressure drop of a single air channel results to 9 mbar. The total pressure drop in the cassette, including internal flow distribution and manifold, results to 28 mbar.

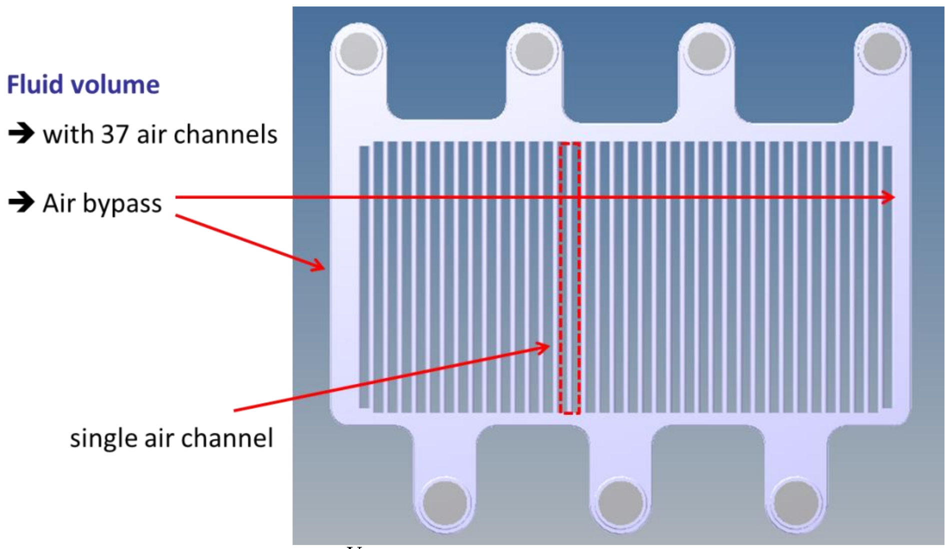

Figure 6. Design CS^V: Fluid volume of the cathode side.

Having determined the flow resistance of the single layers of the cathode side the calculation of the flow distribution to the single layers in a 55 layer stack was performed. First of all a simplification of the stack geometry has to be done to limit the mesh size and so the calculation time. The simplified model of a stack with 55 layers with the resulting pressure drop distribution with non-optimized geometry is shown in Fig. 7.

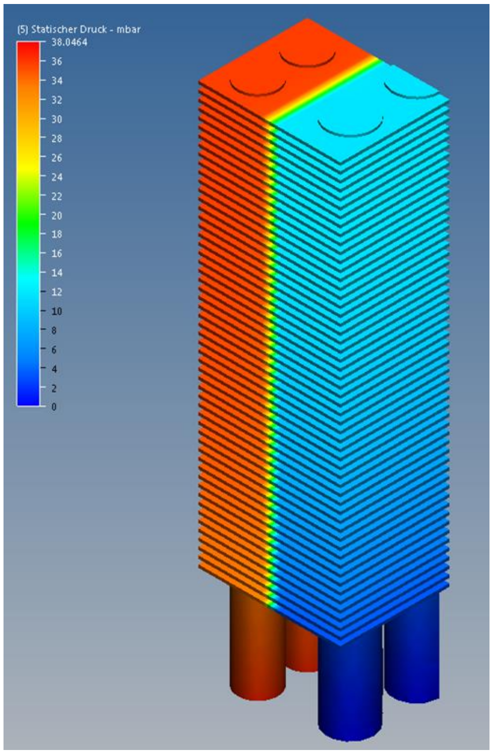

Figure 7. Design CS^V: Simplified model of a stack with 55 layers (non-optimized).

Using the manifold geometries of the first design (see Fig. 4) the flow to the various layers of the stack varies by +10% to -6% (see Fig. 8). By increasing the outlet manifold diameters by about 20% the distribution varies by +4% to -2% (see Fig. 8) which is below the set limit of ±5%.

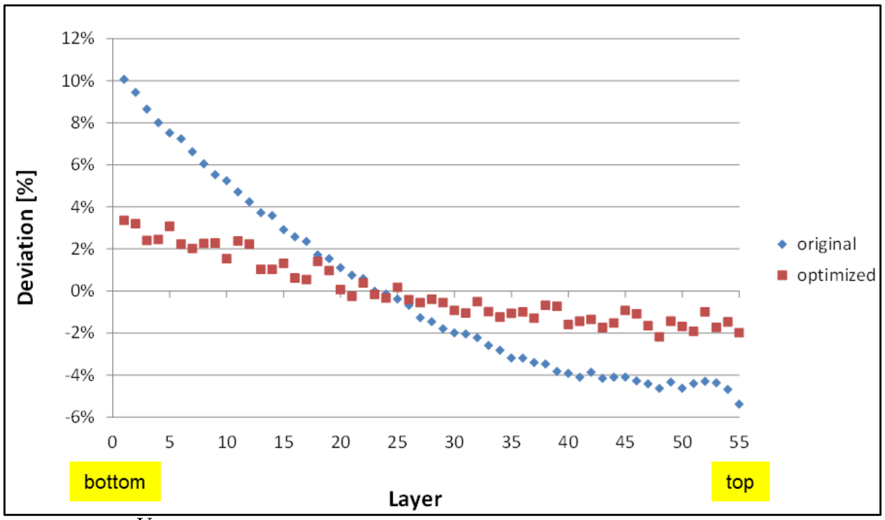

Figure 8. Design CS^V: Flow distribution of the cathode side of a stack with 55 layers.

A comparable calculation war performed for the anode side. Because of the much lower mass flow and the lower gas density here the pressure drop in one layer results to only 2.5 mbar. Using again the manifold geometries of figure 4 the flow to the 55 layers of the stack varies by +19% to -9% (see Fig. 9). By increasing the outlet manifold diameters by about 30% the distribution is very much improved and varies now by +5% to -2% (see Fig. 9) which is below the set limit of ±5%.

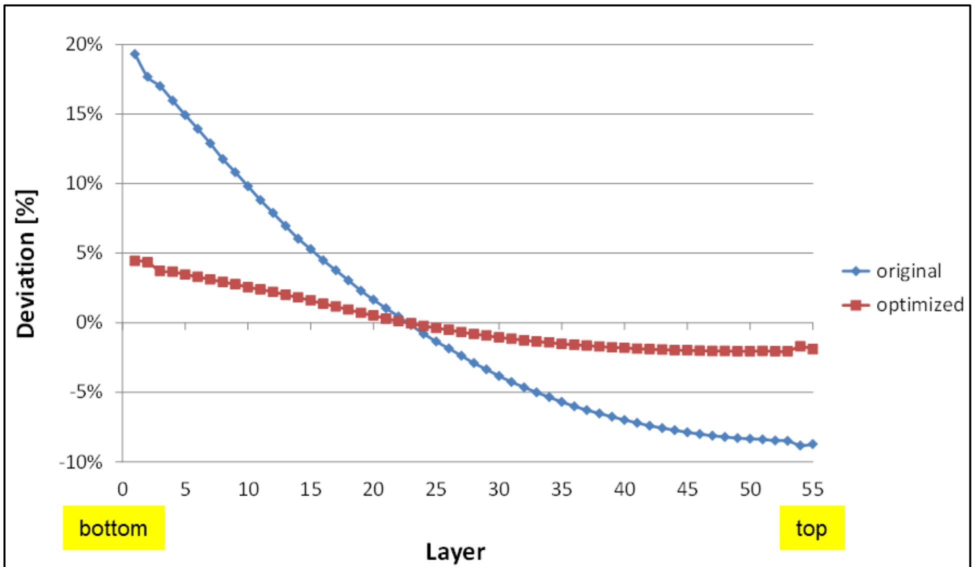

Figure 9. Design CS^V: Flow distribution of the anode side of a stack with 55 layers.

The design CS^V generated from the above described calculations and simulations was further optimized under manufacturing aspects in several iteration steps with Borit's Hydrogate™ forming process.

The final cell and stack dimensions of the new CS^V design are given in the table below.

TABLE I. Final CS^V cell and stack dimensions.

Part	Dimension
Cell dimensions	180 mm x 100 mm
Cell area	180 cm²
Cell thickness	380 µm ± 20µm
Cathode dimensions	160 mm x 80 mm
Cathode area	128 cm²
Stack foot print	209 mm x 195 mm

Manufacturing

The single repeating units (SRU's) of the stack, the trays and the frames have been manufactured by Borit's Hydrogate™ process, which is based on continuous hydroforming. Utilizing hydrostatic pressure, Hydrogate™ is able to produce challenging geometries with low residual stress resulting in very flat plates. As a single step process, Hydrogate™ offers repeatable quality, extreme flexibility, low tooling costs and high productivity [7]. Details of the process are shown in Figures 10, 11 and 12.

Figure 10. Advantages of the Hydrogate™ process of Borit [7].

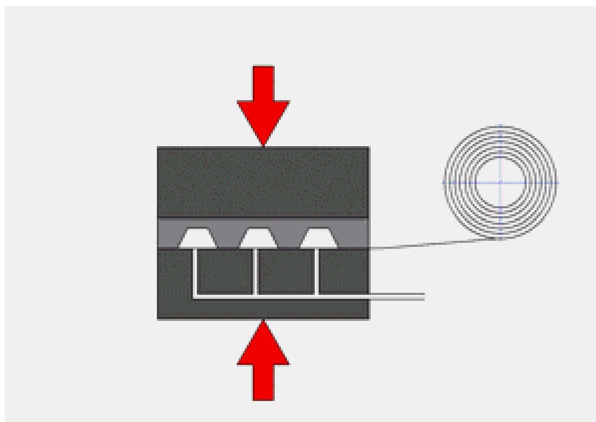

Figure 11. Hydrogate™ Process Principle by Borit [7].

The manufacturing process is set-up suitable for mass production and cost effectiveness by minimizing manufacturing and handling processes.

Figure 12. Manufacturing process of SRU's metal sheet parts, tray and frame.

Crofer 22 H is used as material for the SRU's with 0.3 mm thickness. The high-chromium ferritic steel, which was specially designed for SOFC interconnect applications, was mainly optimized with respect to low oxidation rates, high electrical conductivity of the surface oxide scales, low Cr evaporation and suitable workability [8, 9]. These properties were obtained by defining very low concentrations for minor alloying additions such as silicon and aluminum commonly present in the steel. The material showed excellent behavior during the hydroforming process. Non visual defects i.e. cracks have been observed so far.

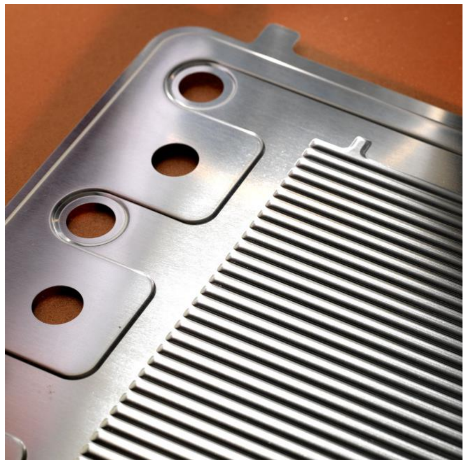

Figure 13. SRU part (tray) in CS^V design manufactured by Borit.

Assembly and Sealing

With the aim of developing processes suitable for industrial production, Forschungszentrum Jülich is working on two application processes of the glass ceramic sealant besides the standard application directly to the stack parts by a dispenser. A first alternative application process is screen printing of the glass ceramic sealant directly to the stack parts. A second method, developed by Forschungszentrum Jülich, is a type of net-shape gasket where the glass-ceramic sealant is applied and dried on a foil (Fig. 14). The dried sealant is removed from the foil and placed on the stack parts. Its essential advantages are first the decoupling of the sealant manufacturing from stack manufacturing and secondly the sealant on foil is storable. Thus the concept of the sealant on foil provides an enormous manufacturing flexibility [9, 10].

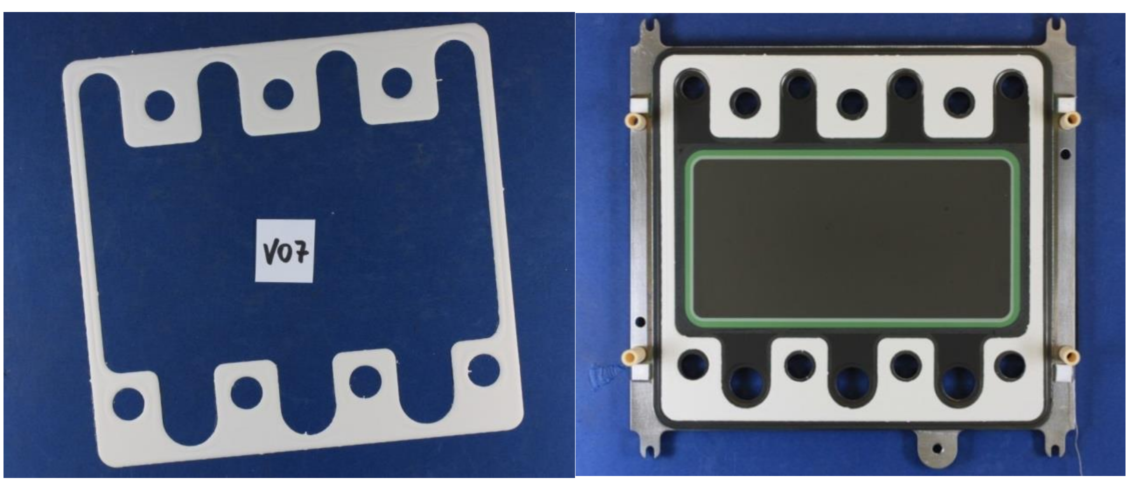

Figure 14. Left side: Glass ceramic sealant on foil. Right side: sealant on cassette.

Composites prepared from a glass matrix based on the BaO-CaO-SiO_2 system and YSZ fibers as a filler additive [12] are used as sealing material.

While the first development steps were mainly focused on adapting thermal expansion and chemical stability, a sufficient mechanical strength of the sealants was shown to be the major critical task in the recent years [5]. The temperature gradients occurring during stack operation can be critical for the joints, which need to withstand the resulting thermal stresses. The composite material with YSZ fibers showed the best results with respect to mechanical strength [13, 14]. A high reproducibility of gas-tight sealings was demonstrated over the past year in several SOFC stacks using the glass-ceramic composite sealant H with 13 wt.-% YSZ fibers in combination with the interconnect materials.

So far, in all CS^V stacks sealants on foil have been used for joining the cassettes. The three stacks tested until now showed a very good tightness during and after operation (see the test results below).

Testing of CS^V stacks

The functionalities of the optimized components and design variations were tested first with a two-layer short stack (stack number: CS^V02-01). The joining process was carried out in a furnace at 850 °C, with a clamping weight of 75 kg. After cell reduction at 800 °C by increasing the hydrogen concentration stepwise, the stack performance was characterized by voltage-current (U-j curve) measurements at furnace temperatures of 700 °C, 750 °C and 800 °C. No stationary operation was carried out with stack CS^V02-01 due to its relatively poor performance. The stack was cooled down after characterization for post-test analysis, where it turned out that the screen printed glass sealant tapes were too thick which resulted in a bad electrical contact between cell and interconnect. All other components worked properly. Therefore, the second CS^V stack was assembled directly with five layers (stack number: CS^V05-01), with optimized glass thickness. Joining process and characterization procedure were the same as with CS^V02-01.

The open circuit voltages (OCVs) measured under dry hydrogen can to a certain extent indicate the gas tightness of the stack. The OCVs of both CS^V stacks after reduction are shown in Figure 15. The OCVs were measured at 800 °C with a mixture of H_2 and Ar (H_2: Ar=1:1). The worst cell voltage was 1.181 V, which still corresponds to a steam content of only 0.28%. The good gas tightness was also confirmed by a leakage test at room temperature after the stack was dismounted from the test bench.

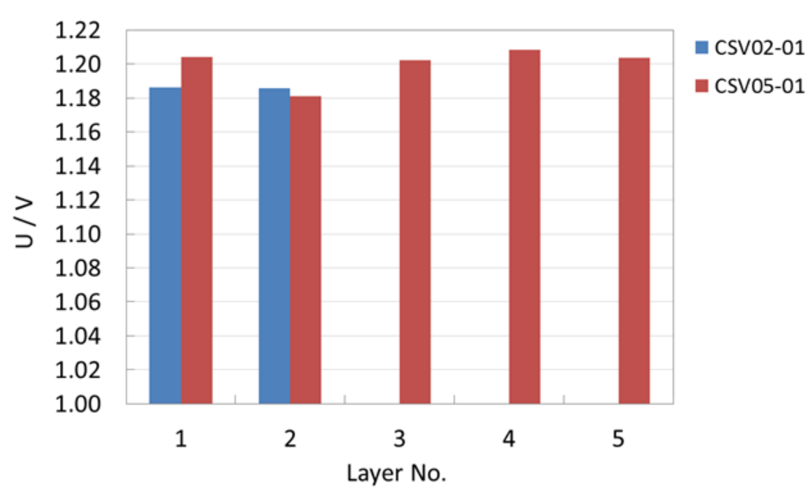

Figure 15. OCVs of both CS^V stacks, measured at 800 °C with dry gas mixture of H_2 and Ar (H_2: Ar=1:1).

The U-j curves of stack CS^V05-01 measured with 10% humidified gas mixture (H_2: Ar=1:1) at three temperatures are shown in Figure 16. The average area specific resistances at each temperature were calculated with the cell voltages at 0.5 Acm^{-2}, and the Arrhenius plots of the two CS^V stacks were compared with one of the best CS^{IV} stacks (CS^{IV}04-05), as shown in Figure 17. After optimization in thickness of the screen printed glass tapes, the second CS^V stack (CS^V05-01) showed comparable or slightly better performance than CS^{IV}04-05, indicating that the newly developed CS^V–design stacks with enlarged cassette and active cell areas can be well produced and assembled under the current manufacturing standard.

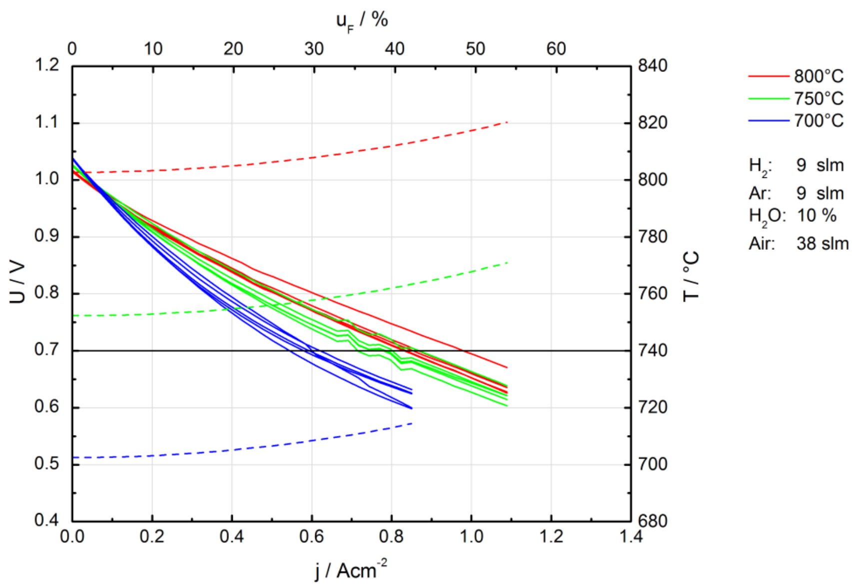

Figure 16. U-j curves of stack CS^V05-01, measured at furnace temperatures of 800 °C, 750 °C and 700 °C with 10% humidified gas mixture (H_2: Ar=1:1).

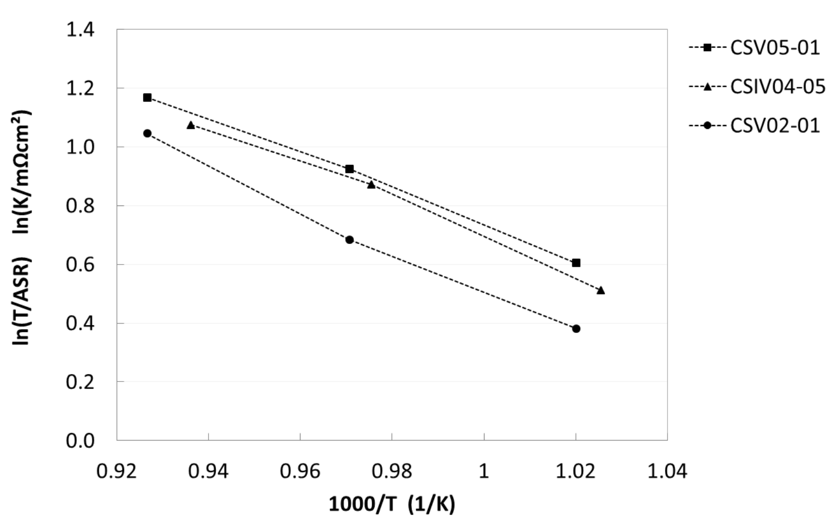

Figure 17. Arrhenius plots of the two CS^V stacks and one CS^{IV} stack

Further tests will be focused on production reproducibility, thermomechanical stability and long-term stability, also with stacks with an increased number of layers.

Summary and Outlook

The new light weight SOFC design CS^V addresses an improved design solution. It is designed to reduce the high thermical gradients in cassettes and to optimize the fluid dynamic in the cassettes and the whole stack. Furthermore the design is improved regarding the manufacturing and the assembly processes. At the same time the performance of the new CS^V design has shown comparable or even slightly better performance than the previous design, indicating that the newly developed design with enlarged cassette and active cell area can be well produced and assembled under the current manufacturing standards. Summarizing, the new developed CS^V design is able to meet the challenges regarding the requirements for (mechanical) robustness and the manufacturing costs of light weight SOFCs.

The next steps will be tests to prove the thermomechanical robustness by extensive thermal cycling and tests to prove long term stability. Furthermore bigger stacks will be build up with 30 and 60 layers. The design will be further optimized. Ideas for further improvements are already available. One improvement will be the replacement of the inlays, currently made by several parts by only one, reducing further manufacturing costs and material consumption.

Acknowledgments

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